$$\psi_{n+1}(x) = [\ \psi_n(x) * p_{n+1}(x)\ ] \bullet q_{n+1}(x) \qquad \text{(A3)}$$

In this formulation the real-space wave function yn(x) is convolved by a real-space propagator $p_{n+1}(x)$ and the result multiplied by the real-space phase-grating. Van Dyck (1983) has developed this method and shown that the convolution step need not be carried out with the full $2^n x 2^m$ p(x) array, but only with a such smaller array (since p(x) is sharply peaked in the forward direction for high energy scattering). Since the modified p(x) array is so much smaller, the computation time should take less time than either of the reciprocal-space multislice (A1) or the FFT multislice (A2); however, tests (e.g. Kilaas and Gronsky, 1983) have shown that the original formulation of the method required more beams and smaller slices to achieve the same precision as either the reciprocal-space multislice or the FFT multislice, resulting in a longer time to produce the wavefield at the same total specimen thickness. Van Dyck and Coene (1984) have since proposed a modified implementation of the real-space multislice into a workable algorithm, with results that approach those produced by the multislice formulations of equations A1 and A2 (Coene and Van Dyck, 1984). Since the p(x) array is much smaller, procedure (A3) does produce some saving in memory over the other multislice formulations.

A.4 Storage requirements

All three formulations of the multislice procedure require the computer (or array processor) to store three different complex arrays corresponding to the phasegrating function in real or reciprocal space (q(x) or Q(k)), the electron wavefield in real or reciprocal space (y(x) or Y(k)), and the propagator function in real or reciprocal space (p(x) or P(k)).

With a reciprocal-space phasegrating Q(k), of 2nx2m terms, a parameter-(n+m) reciprocal-space multislice (equation A1), requires a reciprocal-space wavefield array (or set of diffracted beams), Y(k), of 2n-1x2m-1 terms, and a reciprocal-space propagator array P(k), of 2n-1x2m-1 terms.

An equivalent parameter-(n+m) FFT multislice (equation A2) requires a real-space phase-grating array q(x), of $2^n x 2^m$ terms, an electron wavefield array Ψ(k), of $2^n x 2^m$ terms, and a reciprocal-space propagator array P(k), of $2^{n-1} x 2^{m-1}$ terms.

For a real-space parameter-(n+m) multislice (equation A3), both the ψ(x) and q(x) arrays need to hold $2^n x 2^m$ terms, whereas p(x) may be as small as thirteen (Kilaas & Gronsky, 1983), giving a total requirement of slightly over $2x2^{n+m}$ terms. In implementing the real-space multislice it is important that the $2^n x 2^m$ q(x) array should be formed from a full $2^n x 2^m$ Q(k) terms, whereas the $2^n x 2^m$ y(x) array should be formed from only 2^{n-1} $x2^{m-1}$ Ψ(k) terms in order to include correctly all the physical scattering contributions to each diffracted beam and to avoid aliasing problems (fig A1).

Any (correctly implemented) multislice of size $2^n x 2^m$ (parameter n+m) thus includes the effects of only $2^{n-1} x 2^{m-1}$ diffracted beams. This is true for all three formulations of the multislice: the reciprocal-space method (equation A1), the FFT method (equation A2), and the real-space multislice (equation A3).

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Appendix BHOLZ interactions

With suitable algorithms, it is possible to include in the diffraction calculation the effects of out-of-zone scatterings, or non-zero (or higher-order) Laue zone (HOLZ) interactions. Basically, there are four ways to produce the set of phasegratings (or projected potentials) that describe the "multisliced" crystal. For structures with short repeat distances in the beam direction, the simplest method is to use one slice per unit cell. For structures with large repeats in the beam direction, several methods may be used, three of which rely on sub-dividing the slice into "sub-slices". Any of the four methods can be used in NCEMSS.

B.1 Identical slices with only one sub-slice per unit cell repeat distance

A multislice computation in which every slice is identical contains no information about the variation in structure along the incident beam direction, and includes scattering interactions with only the zero-order Laue zone (ZOLZ) layers. For structures with short repeat distances in the beam direction such a computation is adequate, since the Ewald sphere will not approach the (relatively distant) high-order zones.

B.2 Identical sub-slices with n sub-slices per unit cell repeat distance

For structures with large repeats in the beam direction, a method of sub-dividing the slice is required in order to compute the electron scattering with sufficient accuracy. The simplest, but most approximate method, is to compute the projected potential for the full repeat period then use 1/n of the projected potential to form a phase-grating function that can be applied n times to complete the slice. This method avoids interaction with any "pseudo-upper-layer-line" (Goodman and Moodie, 1974), but ignores real HOLZ layers.

B.3 Sub-slices based on atom positions

An improvement on sub-dividing the projected potential is to sub-divide the unit cell atom positions. In this procedure the list of atom positions within the unit cell is divided into n groups depending upon the atom position in the incident beam direction. From these sub-sliced groups, different projected potentials are produced to form n different phase-gratings, which are applied successively to produce the scattering from the full slice.

B.4 Sub-slices based on the three-dimensional potential

A further improvement on sub-dividing the atom positions, is to sub-divide the three-dimensional potential of the full slice, since an atom with a position within one sub-slice can have a potential field that extends into the next sub-slice. Rather than compute a full three-dimensional potential and then integrate over appropriate sub-slic-

es (a 128x128x128 potential would require over two million samples to be stored), it is possible to derive an analytical expression for the potential within the sub-slice z0 ± Dz projected onto the plane at z0 (Self et al., 1983). It is possible to apply this method routinely to structures with large repeats in the beam direction, thus generating several different phase-gratings for successive application, and even to structures (perhaps with defects) that are aperiodic in the beam direction and require a large number of individual non-repeating phase-gratings (Kilaas et al., 1987).

B.5 NCEMSS sub-slicing

While ensuring that the calculation remains sufficiently accurate, NCEMSS will normally choose the simplest (and quickest) method of specifying how slices are defined for any particular combination of specimen, zone axis, accelerating voltage, and maximum g. To this end, the user can choose to neglect HOLZ interactions if these are judged to be unimportant. If HOLZ interactions are important, then the user should select the "3D-POT.SLICE" box in the SET-UP menu (p 41), rather than the "2D-POT.SLICE" box.

When a two-dimensional calculation is selected, NCEMSS will use one slice per cell if the cell repeat distance in the beam direction is small (B.1). If the repeat distance is too large for one slice per unit cell, NCEMSS will avoid pseudo-upper-layer-lines by producing n identical sub-slices (B.2).

When a three-dimensional calculation is selected, (3D-POT.SLICE activated), NCEMSS uses a sub-divided three-dimensional potential (B.4) when the repeat distance is large, and defaults to one slice per cell if the distance is small enough. Note that the number of sub-slices per unit cell can be forced to be greater than one by setting it explicitly in the MAIN menu (p 16); this will ensure that any HOLZ interactions are included even for small repeat distances. Of course, if the repeat distance is very small, leading to a distant HOLZ in reciprocal space, both the calculation and the experiment it is modeling will interact only very weakly with the HOLZ reflections.

Use of the LAYERED STRUCTURE option (p 12) to produce the scattering from a structure that is layered or aperiodic in the incident beam direction is effectively an application of the method of sub-slicing based on atom positions (B.3). Thus the user could create a number of sub-slices by assigning selected atoms to different structure files, then forming a phasegrating for each sub-slice, and using the SET-LAYERS menu (p 46) to specify how the sub-slices are to be used to describe the specimen structure. Normally, of course, the use of the B.4 option is to be preferred, as it produces a superior result, and is far easier to use.

B.6 Other methods

Van Dyck has proposed other methods to include the effects of HOLZ layers, including the second-order multislice with potential eccentricity (Van Dyck, 1980) and the improved phase-grating method (Van Dyck, 1983). Tests of these procedures show that the extra computation involved in using potential eccentricity may be worthwhile, but that the improved phase-grating method diverges too easily to be useful.

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